

# Universal scaling and quantum critical behavior of $\text{CeRhSb}_{1-x}\text{Sn}_x$

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We propose a universal scaling  $\rho\chi = \text{const}$  of the electrical resistivity  $\rho$  with the inverse magnetic susceptibility  $\chi^{-1}$  below the temperature of the *quantum-coherence onset* for the Ce 4f states in  $\text{CeRhSb}_{1-x}\text{Sn}_x$ . In the regime, where the Kondo gap disappears ( $x \simeq 0.12$ ), the system forms a *non-Fermi liquid* (NFL), which transforms into a Fermi liquid at higher temperature. The NFL behavior is attributed to the presence of a novel *quantum critical point* (QCP) at the Kondo insulator - correlated metal boundary. The divergent behavior of the resistivity, the susceptibility, and the specific heat has been determined when approaching QCP from the metallic side.

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The compounds  $\text{CeRhSb}$  and  $\text{CeRhSn}$ , both with strongly correlated nature of 4f electrons, form unprecedented types of ground state:  $\text{CeRhSb}$  represents the *Kondo semiconductor* with a gap  $\Delta \simeq 7$  K [1] and  $\text{CeRhSn}$  is a *non-Fermi* (non-Landau) quantum liquid [2].

The fundamental question is whether by studying  $\text{CeRhSb}_{1-x}\text{Sn}_x$  the two states can be related through a corresponding *quantum critical point* (QCP) or a phase boundary located in between. In that respect, the quantum critical points between antiferromagnetic metal and either superconducting [3] or paramagnetic [4] heavy-fermion states have been successfully identified play an important role in the discussion of the origin of exotic superconductivity [3]. Also, the existence of the QCP between the Kondo insulator and an itinerant antiferromagnet has been suggested [5] as a function of either the Kondo coupling or pressure.

In this paper we show directly that a transition of this type exhibits indeed a well defined quantum critical behavior as a function of the number of carriers. The number of the carriers per 4f electron is a crucial factor when examining the formation of the collective Kondo-singlet state (cf. famous "exhaustion" theorem of Nozières [6]). To address these basic topics, we have studied the series  $\text{CeRhSb}_{1-x}\text{Sn}_x$ , in which the number of valence electrons diminishes by one per formula when substituting Sn atom for Sb. The characterization of such QCP allows also to compare the insulator-metal transition for the *Mott-Hubbard* systems and that for the *Anderson-Kondo* lattices. We show that the behavior in Ce compounds differs from that for the charge-transfer Mott-Hubbard systems [6], even though the hybridization between the strongly correlated and valence states is common to both classes. It implies that the *Mott-Hubbard* and *Anderson-*

TABLE I: The Kondo gap  $\Delta$  for the  $\text{CeRhSb}_{1-x}\text{Sn}_x$  samples for  $x \leq 0.12$ .

x	$\Delta(K)$
0	6.70
0.01	1.80
0.02	0.64
0.04	0.70
0.06	0.30
0.08	0.22
0.10	0.10
0.12	0.10

*Kondo* lattice systems may belong to separate *universality classes* of systems with quantum phase transitions.

We first demonstrate, on the basis of experimental-data correlation, a novel type of scaling between the electrical resistivity  $\rho$  and the magnetic susceptibility  $\chi$  in the *quantum-coherence regime* for the Kondo semiconductors  $\text{CeRhSb}_{1-x}\text{Sn}_x$ , with  $x \leq 0.12$ . The reference resistivity data are displayed in Fig. 1ab. We are interested in the properties below the temperature  $T_{max}$ , where the resistivity has a maximum. For  $T > T_{max}$  a pronounced  $\ln T$  behavior of  $\rho$  should be noted for all the samples studied [7]. However, for  $T \ll T_{max}$  a pronounced exponential increase of  $\rho \sim \exp(\Delta/T)$ , representing a nondegenerate semiconductor, can be fitted well for  $x < 0.12$ ; the corresponding gap values  $\Delta = \Delta(x)$  are listed in Table I. The properties of the metallic state for  $x > 0.12$  will be discussed below.

In Fig. 2 we show exemplary data (for  $x = 0$ ) of the scaling of  $1/\rho$  with  $\chi$ ; the inset provides the detailed behavior. One should note that in order to obtain such a good scaling, we have to subtract from the measured  $\chi$  the impurity Curie-law contribution ( $nC/T$ ) with  $n$  in the regime  $0.004 \div 0.008$  (depending on the sample), which represents a standard procedure in these and related compounds [8]. Also, then  $\chi \rightarrow 0$  with  $T \rightarrow 0$ , demonstrating that the Kondo semiconductors can be regarded at  $T = 0$  as either a quasiparticle band insulators

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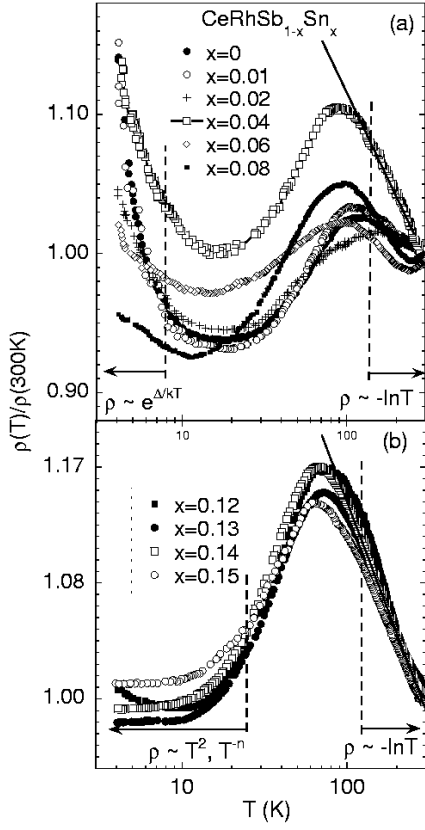


FIG. 1: Temperature dependence of the relative resistivity  $\rho$  for  $\text{CeRhSb}_{1-x}\text{Sn}_x$  systems for  $x \leq 0.08$  (a) and  $0.12 \leq x \leq 0.15$  (b). The regimes, where  $\rho(T) \sim \exp(\Delta/k_B T)$ ,  $\sim -\ln T$ , and  $\sim T^2$  are marked by the corresponding solid lines.

(with a band gap renormalized by the electron correlations [9]) or as a collective Kondo-lattice insulator [10] with a singlet spin state in the ground state. The vanishing  $\chi$  in the ground state distinguishes the Kondo insulators from the Mott-Hubbard insulators, for which the correlated electrons have unpaired spins and thus order magnetically when  $\Delta > 0$ .

The details of the scaling  $\rho\chi = \text{const}(x)$  are demonstrated explicitly in Fig. 3, where the linear dependence between  $\chi$  and  $\rho$  is shown (note that we have coalesced the data to a common origin in each case). The inset illustrates the value of the constant  $R_S = \rho\chi$  as a function of  $x$ . In the regime  $x \lesssim 0.06$  it has a universal value  $R_S \simeq 0.1 \mu\Omega \text{ cm emu/mol}$ . The existence of the scaling displayed in Fig. 3 demonstrates that all the systems exhibit a common universal property, i.e. can be regarded as belonging to a single *class*. It remains to be discussed next whether they form a *phase* in the thermodynamic sense. This can be done only by determining the nature of the boundary separating it from the metallic phase setting in for  $x > 0.12$ .

To characterize the metallic phase we have plotted in Fig. 4 the resistivity data as a function of  $T^2$ . This dependence is well fulfilled for the higher temperatures

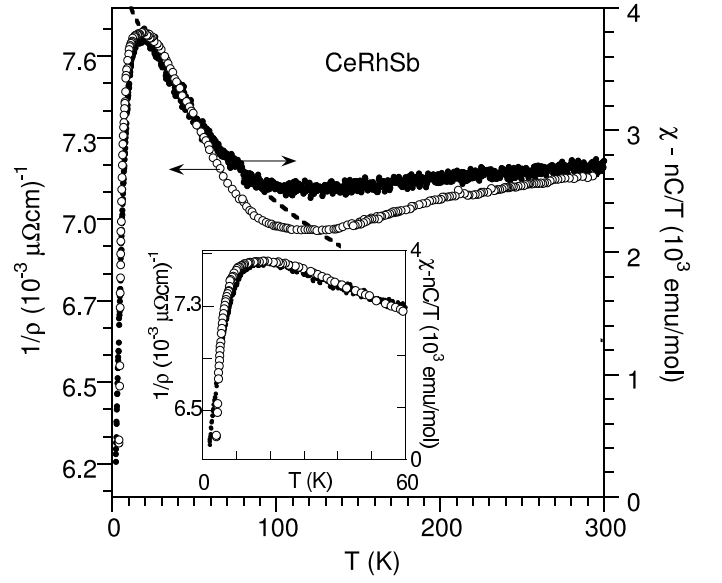


FIG. 2: Temperature dependences of the inverse resistivity  $\rho^{-1}$  (left scale) and of the paramagnetic susceptibility  $\chi$  (with the impurity-contribution  $nC_0/T$ ,  $n = 0.004$ , subtracted) for  $\text{CeRhSb}$ . The dashed line displays the Curie-Weiss contribution to  $\chi$ . The inset:  $\rho^{-1}$  and  $\chi$  on an expanded scale.

$T > 10$  K. In the  $T$  range below about 10 K (see the inset), a clear deviation from the Landau-Pomeranchuk-Baber law  $\rho = \rho_0 + AT^2$  for the Fermi liquid can be observed, particularly for the concentration  $x \simeq 0.13$ , where the gap has barely disappeared. The fitting to the  $\sim T^{-n}$  dependence, with  $n \simeq 0.1$ , should be regarded only as an indicative factor of an incipient *non-Fermi-liquid (NFL) behavior*, requiring an additional test. Alternatively, these low- $T$  data can be represented by the  $\ln T$  dependence.

To determine thermodynamic properties of the non-Fermi liquid we have to discuss the temperature dependence of equilibrium characteristics in the vicinity of the critical point, in our situation placed at  $x \simeq 0.12$  and  $T = 0$ . For that purpose, we have shown in Fig. 5 the temperature dependence of  $\chi(T)$  for  $0.13 \leq x \leq 0.16$ . In each case, a clear dependence  $\chi = aT^{-m}$  is observed. The values of the parameters are listed in Table II. The quantities  $\chi^{-1}$  and  $\rho$  now do not obey the linear type of relation shown in Fig. 3 for KI state. Also, now  $\chi$  diverges, whereas  $\rho$  approaches the finite value  $\rho_0$  when  $T \rightarrow 0$ . The divergence of  $\chi$  signals also a magnetic phase transition, probably to weakly ferromagnetic or Griffiths phases [11]. The onset of magnetism signals a spontaneous symmetry breakdown at  $T = 0$  needed for the *critical point* to be well defined. The onset of magnetism cannot be related to the Kondo insulating phase, since then  $\chi \rightarrow 0$ . So, whereas in Mott-Hubbard systems the insulating phase is magnetic (very often antiferromagnetic) and the metallic phase can be paramagnetic, here the Kondo insulator is nonmagnetic, whereas the metallic phase is magnetic, most probably with a very small

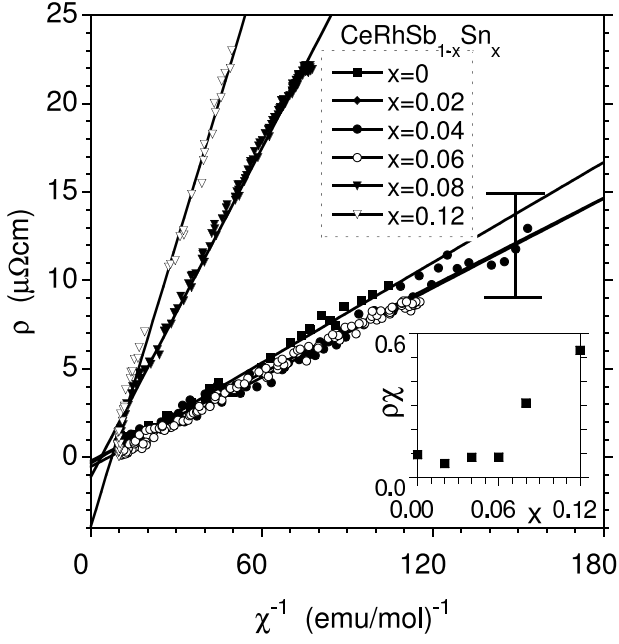


FIG. 3: Linear scaling law between the resistivity  $\rho$  and the susceptibility  $\chi$  for the systems with nonvanishing Kondo gap. The quantities label the corresponding  $\rho$  and  $\chi$  values shifted to the zero value at the minimal positions. The error bars represent the inaccuracy of  $\rho$  measurements.

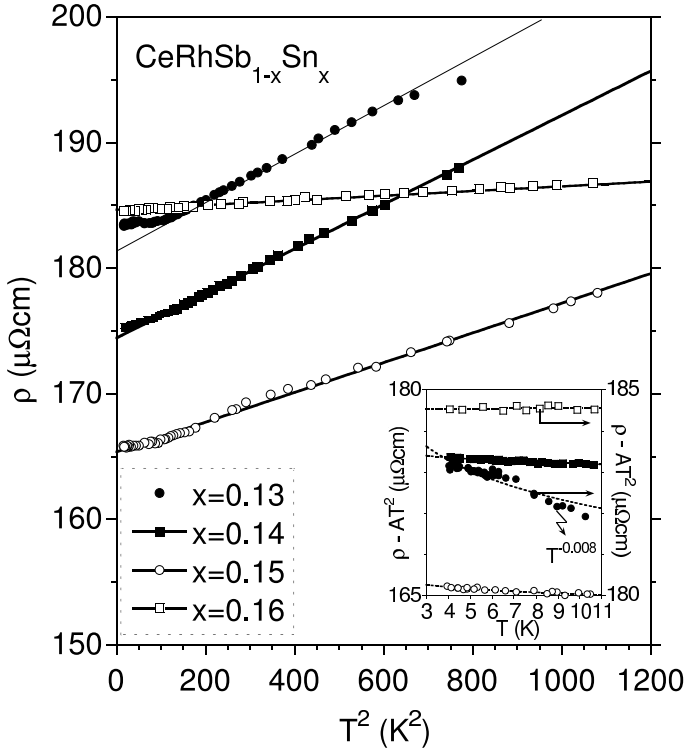


FIG. 4: Resistivity vs.  $T^2$  for the samples with  $0.13 \leq x \leq 0.16$ . The inset: non-Fermi-liquid-type scaling  $T^{-n}$ ,  $n = 0$ , for the sample  $x = 0.13$ , i.e. close to the critical point where the Kondo gap disappears. The lines are the fitted curves.

moment, as in many heavy-fermion systems. The existence of a weakly ferromagnetic phase in Sn-rich samples has been observed earlier [2], so we will not discuss it in detail here.

We have also performed the specific heat ( $C_p$ ) measurements on the sample with  $x = 0.13$ . The data in the temperature range 10 – 20 K can be fitted to the dependence  $C_p/T = \gamma + \beta T^2$ , with  $\gamma = 63.3$  mJ/K<sup>2</sup>mol. Below 10 K the data start deviating from this dependence and the difference can be fitted to the formula  $C_p/T = bT^{-s}$ , with  $b = 143.6$  mJ/K<sup>1.7</sup>, and  $s \approx 0.3$ . So again, as in the case of the resistivity, the NFL critical regime appears for  $T \rightarrow 0$  from the gross Fermi-liquid-like state.

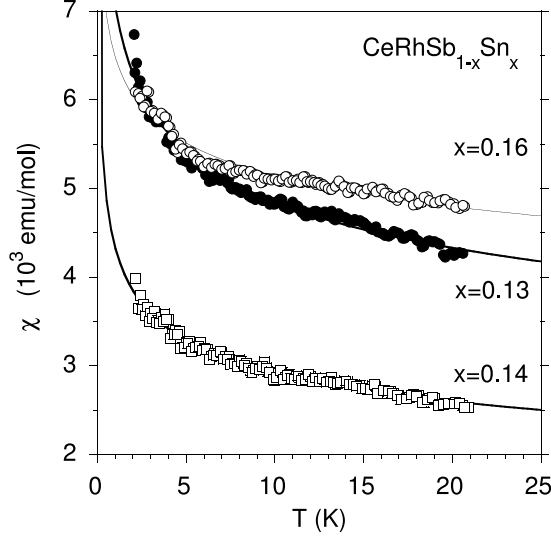
One additional feature of the analysis should be mentioned. The  $\chi(T)$  data for  $T > 15$  K and in the metallic phase ( $x \geq 0.13$ ) can be parametrized in the form:  $\chi(T) = \chi_0 + C/(T - \Theta)$ , where value of the  $\chi_0 \approx 1.7 \div 3.5 \cdot 10^{-3}$  emu/mol, a value in the range for the heavy fermions (cf. Table II).

We now discuss the results in a broader scope. First, the activated behavior of the electronic conductivity comes from the circumstance that the carrier concentration  $n_c$  for  $x \leq 0.12$  increases by thermal excitation over the Kondo gap  $\Delta$ . The dependence  $\rho \sim \exp(\Delta/T)$  is obeyed even for temperature sizably higher than  $\Delta$  and is in agreement with the well known fact that the Kondo semiconductors represent the systems with a low carrier concentration. Likewise, the magnetic susceptibility  $\chi \sim n_c$ , as the carriers are activated from the singlet spin-paired ground-state configuration with  $\chi \approx 0$ . However, it is important to note that those carriers are coupled antiferromagnetically, as the susceptibility exhibits the Curie-Weiss behavior (cf. Fig. 3) with a sizable paramagnetic Curie temperature  $\Theta \approx -124$  K, and the value of the Curie constant  $C = 0.5$  emu K/mol, the value of which is about a half of that for the free Ce<sup>3+</sup> ion. Theoretically, up to a half of the 4f moment compensation for the Kondo lattice comes from the carriers [9]. So, the Kondo gap has a magnetic origin. Second, the system remains Kondo insulating even by doping CeRhSb with Sn. This means that the interpretation of the Kondo insulating state as a hybridized-band insulator is not quite accurate, since it represents a *collective non-magnetic Kondo-lattice* state, which cannot be formed if the number of valence electrons is diminished below a critical value. However, the situation is not easy because of the atomic disorder introduced with the substitution, the role of which has also been discussed in these low carrier-density systems [12]. Nevertheless, the role of disorder does not seem to be a decisive factor in the formation of the Kondo insulating state, since the gap drops with the increasing  $x$ . The residual value of the gap  $\Delta \approx 0.1$  K for the samples with  $x = 0.10$  and  $0.12$  may be due to the disorder. This factor may, however, broaden the critical regime to certain interval around  $x \approx 0.12$ , as is seen in experiment, since the critical behavior of  $\chi$  is seen even for  $x = 0.16$  (cf. Fig. 5).

In conclusion, we have shown that the Kondo semicon-

TABLE II: Susceptibility and the resistivity parametrizations of the samples in the *metallic* regime ( $x \geq 0.13$ ).

x	$\chi = \chi_0 + C/(T - \Theta)$			$\chi = aT^{-m}$		$\rho = \rho_0 + AT^2$	$\rho - AT^2 \sim T^{-n}$
	$\chi_0$ ( $10^3 \frac{\text{emu}}{\text{mol}}$ )	$\Theta$ (K)	$C$ ( $\frac{\text{emu K}}{\text{mol}}$ )	$10^3 a$	$m$	$A(10^2 \mu\Omega \text{ cm K}^2)$	$10^3 n$
0.13	2.6	-7.0	0.043	6.0	0.17	1.78	8.0
0.14	1.7	-9.1	0.021	4.3	0.17	1.69	2.8
0.16	3.5	-7.6	0.038	6.0	0.09	0.186	1.3
$15 \text{ K} \leq T \leq 90 \text{ K}$				$T < 10 \text{ K}$		$30 \text{ K} \geq T \geq 10 \text{ K}$	$T < 10 \text{ K}$

FIG. 5: Magnetic susceptibility for  $T < 10 \text{ K}$  and the fitted scaling  $\sim T^{-m}$  (see main text).

ductors  $\text{CeRhSb}_{1-x}\text{Sn}_x$ , with  $x \leq 0.12$  are characterized

not only by the gap in the conductivity and the vanishing paramagnetic susceptibility for  $T \rightarrow 0$ , but also by the universal scaling law  $\rho\chi = \text{const}(x)$ . Also, the system undergoes the nonmagnetic Kondo insulator-metal transition at  $x \approx 0.12$ . On the metallic side, a novel quantum critical point and NFL behavior has been discovered for the samples  $x \rightarrow 0.12+$ , that is specified by the power-law increase ( $T^{-\alpha}$ ) of the resistivity, the susceptibility, and the specific heat at lower temperatures. The simultaneous divergences of both  $\rho$  and  $\chi$  at QCP illustrates nicely the circumstance that both the onset of the KI gap and the magnetic critical fluctuations appearance coexist then. The transition is markedly different from the Mott-Hubbard transition. The non-Fermi liquid behavior emerges with  $T \rightarrow 0$  from an overall Fermi-liquid state. Also, the quantum critical behavior described here as a function of the carrier number differs from that appearing [13] at the paramagnetic heavy fermion - antiferromagnetic metal boundary.

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